TH

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
10/798,598	03/10/2004	Min-Hon Rei	DF-04500	6354

7590 08/17/2007

Haverstock & Owens LLP
162 North Wolfe Road
Sunnyvale, CA 94086

EXAMINER	
BAND, MICHAEL A	
ART UNIT	**PAPER NUMBER**
1753	

MAIL DATE	DELIVERY MODE
08/17/2007	PAPER

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Office Action Summary	Application No.	Applicant(s)
	10/798,598	REI ET AL.
	Examiner	Art Unit
	Michael Band	1753

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☒ Responsive to communication(s) filed on *10 March 2004*.

2a)☐ This action is **FINAL**. 2b)☒ This action is non-final.

3)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims

4)☒ Claim(s) *1-33* is/are pending in the application.

4a) Of the above claim(s) _____ is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☒ Claim(s) *1,2,4,5,12,14,15,17-19,21,22,27,29,32 and 33* is/are rejected.

7)☒ Claim(s) *3,6-11,13,16,20,23-26,28,30 and 31* is/are objected to.

8)☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

9)☒ The specification is objected to by the Examiner.

10)☒ The drawing(s) filed on *10 March 2004* is/are: a)☒ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

Priority under 35 U.S.C. § 119

12)☒ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

a)☒ All b)☐ Some * c)☐ None of:

1.☒ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. _____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

* See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☒ Notice of References Cited (PTO-892)
2) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☒ Information Disclosure Statement(s) (PTO/SB/08) Paper No(s)/Mail Date *3/10/2004*.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application
6) ☐ Other: _____.

DETAILED ACTION

Specification

1. The disclosure is objected to because of the following informalities: "8~10 N HCl" (p. 4, para 14) and "10N HCl" (p. 8, para 49; p. 9, para 52). This should read 8-10 M (molar) HCl. Appropriate correction is required.

Claim Objections

2. Claims 3 and 20 objected to because of the following informalities: "8-10 N HCl". This should read 8-10 M (molar) HCl. Appropriate correction is required.

Claim Rejections - 35 USC § 102

3. The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

> A person shall be entitled to a patent unless –
>
> (b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

4. Claim 17 is rejected under 35 U.S.C. 102(b) as being anticipated by Lee et al (US Patent No. 6,379,524).

With respect to claim 17, Lee '524 discloses a process for manufacturing a composite membrane for separation of hydrogen gas using palladium (abstract). Lee '524 further discloses examples of plating methods designed to prepare a palladium composite membrane by means of coating (i.e. filling) palladium metal membrane and

palladium alloy membrane on a porous support by electroless plating (col. 2, lines 17-20), with a porous support derived from stainless steel (col. 2, lines 11-13). It is known that electroless plating a palladium metal involves using a palladium salt solution, as evidenced by Nogami et al (US Patent No. 6,059,940; col. 1, lines 29-33).

Claim Rejections - 35 USC § 103

5. The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

> (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

6. Claims 1-2, 4-5, and 15 are rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846) in view of Lee et al (US Patent No. 6,379,524).

With respect to claim 1, Makrides '846 depicts a hydrogen diffusion membrane (figure 1) with a metal (part 2) composed of Group 5 elements (vanadium, tantalum, and niobium) with two layers of palladium (parts 4 and 6). Part 2 is etched (i.e. filled), where the palladium is sputtered onto both sides to form films part 4 and part 6 (col. 3, lines 30-39). It is well known that DC sputtering is a preferred method for sputtering conductive metals (i.e. palladium) as evidenced by Hariu et al (Document U of PTO-892, p. 1, filed 06-21-2007). Makrides '846 is also limiting in that while both palladium

films are deposited via sputtering or evaporation (col. 3, lines 46-48), it is not specifically suggested to use electroless plating for one of the palladium films.

Lee '524 teaches a process for manufacturing a composite membrane for separation of hydrogen gas using palladium, which employs the step of electroplating under vacuum an alloy of a palladium compound and a transition metal (abstract), where the transition metal is a Group VA element, such as Ta (tantalum), Nb (niobium), and V (vanadium) (col. 3, lines 59-64). Furthermore, Lee '524 discusses plating methods designed to prepare a palladium composite membrane by means of coating palladium metal membrane and palladium alloy membrane on a porous support includes electroless plating (col. 2, lines 17-20). Lee '524 also states a manufacturing example of a plating solution (i.e. electroless plating) derived from palladium alloy (col. 5, lines 35-60). It is known that electroless plating a palladium metal involves using a palladium salt solution, as evidenced by Nogami et al (US Patent No. 6,059,940; col. 1, lines 29-33). Lee '524 cites the advantages of using an electroless plating solution and a porous support as a composite membrane with better permeation and separation coefficient (col. 1, lines 9-11).

Electroless plating is known to encompass a chemical reaction in an aqueous solution, where hydrogen is released (i.e. evaporated) as evidenced by www.wikipedia.com (Document U of PTO-892, p. 2, filed 06-21-2007). Since Makrides '846 depicts two different palladium layers (figure 1, parts 4 and 6) and different methods of depositing the palladium layers (sputtering and evaporation; col. 3, lines 46-48), it would have been obvious to one of ordinary skill in the art to deposit one layer via

electroless plating as suggested by Lee '524 and deposit the other (i.e. additional) layer via sputtering described by Makrides '846 in order to gain the advantages of better permeation and separation coefficient.

With respect to claim 2, Makrides '846 is limited in that while it is obvious to use some type of platform or support to sputter onto, it is not mentioned.

Lee '524 further teaches that "some porous support materials according to this invention may include conductive metals such as stainless steel" (col. 4, lines 29-31). Lee '524 cites the advantages of a porous stainless steel support as a lower material cost, little occurrence of corrosion or crack, easier processing, and higher mechanical strength for modulation (col. 2, lines 4-16).

It would have been obvious to one of ordinary skill in the art to use the porous stainless steel support taught in Lee '524 for the invention of Makrides '846 in order to gain the advantages of a lower material cost, little occurrence of corrosion or crack, easier processing, and higher mechanical strength for modulation.

With respect to claims 4 and 5, Makrides '846 further depicts figure 1 with part 2 being a tantalum foil (col. 3, lines 32-33). Makrides '846 also suggests that tantalum and niobium are interchangeable (col. 2, lines 70-72; col. 3, lines 1-3). Furthermore it is well known that tantalum, niobium, and palladium are commercially available as either foils or powders (www.webelements.com, Documents V, W, X of PTO-892, p. 1, filed 06-21-2007).

With respect to claim 15, modified Makrides '846 further discloses that the metals from Group V-B (Nb, Ta, and V) have a thickness of 0.001 inch (25.4 μm) and the

palladium films have a thickness each of 1000 angstroms (0.1 μm) (col. 4, lines 30-35), giving a total palladium membrane thickness of 25.4 μm.

7. Claims 3 and 7-8 are rejected under 35 U.S.C. 103(a) as being unpatentable over Markides et al (US Patent No. 3,350,846) and Lee et el (US Patent No. 6,379,524) as applied to claims 1 and 2 above, and further in view of Steinberg et al (US Patent No. 4,055,686).

With respect to claims 3 and 7-8, the references are cited as discussed for claims 1 and 2. Modified Lee '524 further teaches methods of prepping (i.e. activating) a stainless steel support in a temperature range of 4°C (col. 5, lines 40-45) and 800°C (col. 6, lines 10-15). However modified Lee '524 is limited in that while it is discussed to activate the support, it is not suggested to polish the substrate holder.

Steinberg '686 teaches a method of forming metal hydride films on a surface of a substrate that rests on a stainless steel shroud which is on top of the substrate holder (abstract; col. 2, lines 65-67). Steinberg '686 further teaches that metal hydride films are used in hydrogen diffusion applications (col. 1, lines 33-36). Polishing of the substrate, and thus stainless steel shroud, is by mechanically polishing with number 600 emery paper (col. 4, lines 43-45) followed by cleaning with ultrasonic bath (col. 4, lines 58-59). Etching (i.e. acid-washing) with 10% HCl aqueous solution is also discussed (col. 3, lines 52-53) along with sputter etching (i.e. electro-polishing) via DC and radio frequency sputter etching after filling the substrate, and therefore support, with a metal hydride (col. 2, lines 29-59). Steinberg '686 cites the advantage of cleaning as providing a substrate system tolerant to a difference between the coefficients of expansion of the

film and substrate due to a strong bond between the film and substrate (col. 2, lines 12-19).

It would have been obvious to one of ordinary skill in the art to use the polishing and cleaning methods taught in Steinberg '686 for modified Lee '524 in order to gain the advantages of tolerance in differences of coefficients of expansion between the film and substrate.

8. Claim 6 is rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846) and Lee et al (US Patent No. 6,379,524) as applied to claim 5 above, and further in view of Kimura (US Patent No. 4,574,056).

With respect to claim 6, the references are cited as discussed for claim 5. However modified Makrides '846 is limited in that it is not suggested to mix the metal powder with a palladium paste and epoxy resin.

Kimura '056 teaches electroconductive pastes that are used to bond semiconductor elements (i.e. metals) onto substrates (abstract; col. 1, lines 18-21). Furthermore, Kimura '056 discusses electroconductive pastes known in the art include those produced by blending fine electroconductive powders of palladium and heat-resistant binders such as epoxy resin (col. 2, lines 58-63). Kimura '056 cites the advantage of using this paste as not requiring any undercoat electrode treatment (col. 1, lines 34-37).

It would have been obvious to one of ordinary skill in the art to use the palladium paste and epoxy resin taught in Kimura '056 for the membrane of modified Makrides

'846 in order to gain the advantage of not requiring an undercoat electrode treatment for the substrate and layers.

9. Claim 9 is rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846) and Lee et al (US Patent No. 6,379,524) as applied to claim 1 above, and further in view of Harkness (US Patent No. 6,265,086).

With respect to claim 9, the references are cited as discussed for claim 1. However modified Makrides '846 is limited in that while it is disclosed that a silver-palladium alloy is electroless plated, the salt solution is not specified.

Harkness '086 teaches a method for selective electroless metal deposition on a substrate (abstract). Harkness '086 further teaches suitable sources of metal ions include salts of metals, such as palladium acetate, with a concentration in the range of about 0.001 to about 10 weight percent (col. 6, lines 20-29). Furthermore, Harkness '086 states that "electroless plating solutions also often contain solvents for the metal salt, suitable reducing agents, bases, complexing agents to solubilize the metal salt and special additives to control the solution stability and plating rate (col. 6, lines 30-33). Harkness '086 suggests a reducing agent of hydrazine (NH_2NH_2) with a concentration range of about 0.01 to about 10 weight percent (col. 6, lines 38-39), a base of ammonium hydroxide (NH_4OH) with a concentration of 0.01 to about 10 weight percent (col. 6, lines 45-46), and a complexing agent of EDTA with a concentration range of about 0.01 to about 10 weight percent (col. 6, lines 52-54).

It has been held that differences in concentration or temperature will not support patentability of subject matter encompassed by the prior art unless there is evidence

indicating such concentration or temperature is critical. *In re Aller*, 220 F.2d 454, 456, 105 USPQ 233, 235 (CCPA 1955).

It would have been obvious to one of ordinary skill in the art to use a solution of compounds taught by Harkness '086 for the solution of modified Makrides '846 in order to gain the advantages of controlling the solution stability and plating rate.

10. Claims 10-11 are rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846) and Lee et al (US Patent No. 6,379,524) as applied to claim 1 above, and further in view of Stevens et al (USPGPub 2003/0141018).

With respect to claims 10-11, Makrides '846 depicts a hydrogen diffusion membrane (figure 1) with a metal (part 2) composed of Group 5 elements (vanadium, tantalum, and niobium) with two layers of palladium (parts 4 and 6). Part 2 is etched (i.e. filled), where the palladium is sputtered onto both sides to form flims part 4 and part 6 (col. 3, lines 30-39). It is well known that DC sputtering is a preferred method for sputtering conductive metals (i.e. palladium) as evidenced by Hariu et al (Document U of PTO-892, p. 1, filed 06-21-2007). Makrides '846 further discloses that a palladium-silver alloy may be used for the membrane (col. 5, lines 28-35), thus a palladium membrane and a silver membrane is formed. However Makrides '846 is limiting in that while both palladium films are deposited via sputtering or evaporation (col. 3, lines 46-48), it is not specifically suggested to use electroless plating.

Lee '524 teaches a process for manufacturing a composite membrane for separation of hydrogen gas using palladium, which employs the step of electroplating

under vacuum an alloy of a palladium compound and a transition metal (abstract), where the transition metal is a Group VA element, such as Ta (tantalum), Nb (niobium), vanadium (V), and silver (Ag) (col. 3, lines 59-64). Furthermore, Lee '524 discusses plating methods designed to prepare a palladium composite membrane by means of coating palladium metal membrane and palladium alloy membrane on a porous support includes electroless plating (col. 2, lines 17-20). Lee '524 also states a manufacturing example of a plating solution (i.e. electroless plating) derived from palladium alloy (col. 5, lines 35-60). It is known that electroless plating a palladium metal involves using a palladium salt solution, as evidenced by Nogami et al (US Patent No. 6,059,940; col. 1, lines 29-33). Lee '524 further describes that "some porous support materials according to this invention may include conductive metals such as stainless steel" (col. 4, lines 29-31). Lee'524 also states that electroless plating takes 30 minutes (col. 5, lines 40-45), with an additional 2 hours (120 minutes) of deposition time used in example 2 (col. 6, lines 10-25). Lee '524 cites the advantages as a composite membrane with better permeation and separation coefficient (col. 1, lines 9-11), lower material cost, little occurrence of corrosion or crack, easier processing, and higher mechanical strength for modulation (col. 2, lines 13-16).

Electroless plating is known to encompass a chemical reaction in an aqueous solution, where hydrogen is released (i.e. evaporated) as evidenced by www.wikipedia.com (Document U of PTO-892, p. 2, filed 06-21-2007. Since Makrides '846 depicts two different palladium layers (figure 1, parts 4 and 6) and different methods of depositing the palladium layers (sputtering and evaporation; col. 3, lines 46-

48), it would have been obvious to one of ordinary skill in the art to deposit one layer via electroless plating as suggested by Lee '524 and deposit the other (i.e. additional) layer via sputtering described by Makrides '846 in order to gain the advantages of better permeation and separation coefficient.

In addition, Makrides '846 is limiting in that a specific temperature is not suggested for the electroless plating.

Stevens '018 teaches an electroless deposition apparatus comprising at least one metal selected from the group consisting of noble metals, semi-noble metals, alloys therof (abstract), with examples of noble metals being silver and palladium (p. 2, para 13). Furthermore, Stevens '018 discusses electroless deposition of palladium at a preferred temperature between 20°C and 80°C, with temperature between 40°C and 60°C even more preferred (p. 12, para 0109). Stevens '018 states the advantage of heating as an enhancement of the properties of the deposited materials (p. 10, para 88).

It would have been obvious to one of ordinary skill in the art to use the temperatures taught in Stevens '018 in the modified process of Makrides '846 in order to gain the advantage of enhanced properties of the palladium-silver membrane.

It has been held that in the case where the claimed ranges "overlap or lie inside ranges disclosed by the prior art" a *prima facie* case of obviousness exists. *In re Wertheim*, 541 F.2d 257, 191 USPQ 90 (CCPA 1976).

11. Claim 12 is rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846) and Lee et al (US Patent No. 6,379,524) as

applied to claim 1 above, and further in view of Kulkarni et al (US Patent No. 6,283,357).

With respect to claim 12, the references are cited as discussed for claim 1. However modified Makrides '846 is limited in that while it disclosed that palladium is sputtered and thus a palladium target is used, the palladium purity is not specified.

Kulkarni '357 teaches a method for forming sputter targets (abstract). Kulkarni '357 further teaches that "the sputter target material is a metal, metal oxide, metal silicide or alloy which is to be deposited" onto a substrate, "and is advantageously a highly pure material, preferably having a purity of 99% to 99.99999%" (col. 3, lines 35-38). "These materials include, for example, pure metals, alloys, [silicides], and oxides of tantalum, titanium, tungsten, copper, nickel, chromium, aluminum, cobalt, molybdenum, silver, gold, platinum, ruthenium, rhodium, palladium, iron, bismuth, germanium, niobium, and vanadium (col. 3, lines 38-43).

It would have been obvious to one of ordinary skill in the art to use the palladium sputter target purity taught in Kulkarni '357 for the palladium sputter target of Makrides '846 since Makrides '846 fails to disclose a specific target purity and one of ordinary skill would have a reasonable expectation of success in selecting such a purity.

12. Claim 13 is rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846) and Lee et al (US Patent No. 6,379,524) as applied to claim 1 above, and further in view of Gopalraja et al (USPGPub 2004/0094402).

With respect to claim 13, the references are cited as discussed for claim 1. However modified Makrides '846 is limited in that while sputtering is disclosed, specific vacuum pressures, power input, and temperature are not specified.

Gopalraja '402 teaches a DC magnetron sputtering (abstract) for palladium (Pd) and silver (Ag) deposition materials (p. 3, para 0016) while also suggesting electrochemical plating (i.e. electroless plating) and electroplating (p. 3, para 0021-0022). Furthermore, Gopalraja '402 discusses suitable pressure ranges as 1-30 mTorr or 1-5 mTorr (p. 5, para 0043) accompanied with temperatures between 50 to 70°C (p. 2, para 0014). Gopalraja '402 also discusses a power source capable of biasing a pedestal, where the power range is preferred from 150 to 300 W (p. 5, para 0053). Gopalraja '402 cites the advantage to sputtering using these parameters as confining the plasma generated by capacitive coupling to increase the plasma density, leading to an increased ionization rate in addition to the process being promoted at low pressure (p. 3, para 0024-0025).

It would have been obvious to one of ordinary skill in the art to use the parameters specified for a DC sputtering magnetron taught in Gopalraja '402 for the sputtering apparatus of modified Makrides '846 in order to gain the advantages of superior ionization and improved process at lower pressures.

It has been held that in the case where the claimed ranges "overlap or lie inside ranges disclosed by the prior art" a *prima facie* case of obviousness exists. *In re Wertheim*, 541 F.2d 257, 191 USPQ 90 (CCPA 1976).

13. Claim 14 is rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846) and Lee et al (US Patent No. 6,379,524) as applied to claim 1 above, and further in view of Matsuzaki et al (USPGPub 2004/0238356).

With respect to claim 14, the references are cited as discussed for claim 1. However modified Makrides '846 is limited in that while sputtering the palladium is discussed, the amount of time for sputtering is not specified.

Matsuzaki '356 teaches a silver alloy sputtering target (abstract) with palladium being a component of the alloy (p. 1, para 3). Matsuzaki '356 further teaches a holding time of 0.75 to 3 hours (45 mins to 180 mins) (p. 4, para 48) for DC magnetron sputtering (p. 5, para 69).

It would have been obvious to one of ordinary skill in the art to use the holding time set forth in Matsuzaki '356 for the sputtering time of modified Makrides '846 since modified Makrides '846 fails to disclose a specified time and one of ordinary skill would have a reasonable expectation of success in making the modification.

14. Claim 16 is rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846) and Lee et al (US Patent No. 6,379,524) as applied to claim 1, and in further view of Harris (US Patent No. 4,313,013).

With respect to claim 16, the references are cited as discussed for claim 1. However modified Makrides '846 is limited in that while the membrane is heated, it is not suggested at what temperature, for how long, in what type of atmosphere, or what percentage of hydrogen comprises the gas stream.

Harris '013 discloses a palladium or palladium alloy hydrogen diffusion membrane (abstract) and method of treating (i.e. annealing) the membrane with heat for a specific time in a specific atmosphere (col. 2, lines 53-67). Temperatures of between 350°C and 450°C are used at a time period between 0.1 and 10 hours (col. 2, lines 64-67). Harris '013 also states that hydrogen, argon, or nitrogen gas are used, either static (i.e. atmosphere) or flowing. Furthermore, Harris '013 conducts an example of this with the argon stream being analyzed to determine its content, which is depicted in Table 1 (col. 3, lines 64-68). Table 1 depicts a hydrogen content in the range of 6.6-13.0. Harris '013 cites the advantage of the treatment as a production and recovery of hydrogen from a hydrocarbon source (col. 1, lines 43-47).

It would have been obvious to one of ordinary skill to use the treatment method taught in Harris '013 for the manufacturing method of modified Makrides '846 in order to gain the advantages of production and recovery of hydrogen from a hydrocarbon source.

It has been held that in the case where the claimed ranges "overlap or lie inside ranges disclosed by the prior art" a *prima facie* case of obviousness exists. *In re Wertheim*, 541 F.2d 257, 191 USPQ 90 (CCPA 1976).

15. Claims 18-19, 21-22, 28-29, 32 are rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846) and Lee et al (US Patent No. 6,379,524) in view of Stevens et al (USPGPub 2003/0141018).

With respect to claim 18, Makrides '846 depicts a hydrogen diffusion membrane (figure 1) with a metal (part 2) composed of Group 5 elements (vanadium, tantalum, and

niobium) with two layers of palladium (parts 4 and 6). Part 2 is etched (i.e. filled), where the palladium is sputtered onto both sides to form flims part 4 and part 6 (col. 3, lines 30-39). It is well known that DC sputtering is a preferred method for sputtering conductive metals (i.e. palladium) as evidenced by Hariu et al (Document U of PTO-892, p. 1, filed 06-21-2007). Makrides '846 further discloses that a palladium-silver alloy may be used for the membrane (col. 5, lines 28-35), thus a palladium membrane and a silver membrane is formed. However Makrides '846 is limiting in that while both palladium films are deposited via sputtering or evaporation (col. 3, lines 46-48), it is not specifically suggested to use electroless plating.

Lee '524 teaches a process for manufacturing a composite membrane for separation of hydrogen gas using palladium, which employs the step of electroplating under vacuum an alloy of a palladium compound and a transition metal (abstract), where the transition metal is a Group VA element, such as Ta (tantalum), Nb (niobium), vanadium (V), and silver (Ag) (col. 3, lines 59-64). Furthermore, Lee '524 discusses plating methods designed to prepare a palladium composite membrane by means of coating palladium metal membrane and palladium alloy membrane on a porous support includes electroless plating (col. 2, lines 17-20). Lee '524 also states a manufacturing example of a plating solution (i.e. electroless plating) derived from palladium alloy (col. 5, lines 35-60). It is known that electroless plating a palladium metal involves using a palladium salt solution, as evidenced by Nogami et al (US Patent No. 6,059,940; col. 1, lines 29-33). Lee '524 further describes that "some porous support materials according to this invention may include conductive metals such as stainless steel" (col. 4, lines 29-

31). Lee '524 cites the advantages as a composite membrane with better permeation and separation coefficient (col. 1, lines 9-11), lower material cost, little occurrence of corrosion or crack, easier processing, and higher mechanical strength for modulation (col. 2, lines 13-16).

Electroless plating is known to encompass a chemical reaction in an aqueous solution, where hydrogen is released (i.e. evaporated) as evidenced by www.wikipedia.com (Document U of PTO-892, p. 2, filed 06-21-2007. Since Makrides '846 depicts two different palladium layers (figure 1, parts 4 and 6) and different methods of depositing the palladium layers (sputtering and evaporation; col. 3, lines 46-48), it would have been obvious to one of ordinary skill in the art to deposit one layer via electroless plating as suggested by Lee '524 and deposit the other (i.e. additional) layer via sputtering described by Makrides '846 in order to gain the advantages of better permeation and separation coefficient.

In addition, Makrides '846 is limiting in that while it suggests heating the membrane, it is not disclosed to slowly cooling the membrane afterwards.

Stevens '018 teaches an electroless deposition apparatus comprising at least one metal selected from the group consisting of noble metals, semi-noble metals, alloys therof (abstract), with examples of noble metals being silver and palladium (p. 2, para 13). Furthermore, Stevens '018 discusses that the process of depositing the conductive (i.e. palladium-silver alloy) layer (i.e. membrane) may include annealing the substrate in a thermal anneal chamber. Stevens '018 states the advantage of annealing as an enhancement of the properties of the deposited materials (p. 10, para 88).

It would have been obvious to one of ordinary skill in the art to include an anneal step taught in Stevens '018 in the modified process of Makrides '846 in order to gain the advantage of enhanced properties of the palladium-silver membrane.

With respect to claim 19, Makrides '846 is limited in that while it is obvious to use some type of platform or support to sputter onto, it is not mentioned.

Lee '524 further teaches that "some porous support materials according to this invention may include conductive metals such as stainless steel" (col. 4, lines 29-31). Lee '524 cites the advantages of a porous stainless steel support as a lower material cost, little occurrence of corrosion or crack, easier processing, and higher mechanical strength for modulation (col. 2, lines 4-16).

It would have been obvious to one of ordinary skill in the art to use the porous stainless steel support taught in Lee '524 for the invention of Makrides '846 in order to gain the advantages of a lower material cost, little occurrence of corrosion or crack, easier processing, and higher mechanical strength for modulation.

With respect to claims 21 and 22, modified Makrides '846 further depicts figure 2 with part 2 being a tantalum foil (col. 3, lines 32-33). Makrides '846 also suggests that tantalum and niobium are interchangeable (col. 2, lines 70-72; col. 3, lines 1-3). Furthermore it is well known that tantalum, niobium, and palladium are commercially available as either foils or powders (www.webelements.com, Documents V, W, X of PTO-892, p. 1, filed 06-21-2007).

With respect to claim 28, Stevens further teaches electroless deposition of palladium at a preferred temperature between 20°C and 80°C, with temperature between 40°C and 60°C even more preferred (p. 12, para 0109).

It has been held that in the case where the claimed ranges "overlap or lie inside ranges disclosed by the prior art" a *prima facie* case of obviousness exists. *In re Wertheim*, 541 F.2d 257, 191 USPQ 90 (CCPA 1976).

With respect to claim 29, modified Makrides '846 further discloses "a palladium-silver alloy containing silver in an amount falling within the range of about 10 percent to about 50 percent weight" (col. 5, lines 33-35), leading to a weight percent ratio of palladium/silver of about 90/10 to 50/50.

It has been held that differences in concentration or temperature will not support patentability of subject matter encompassed by the prior art unless there is evidence indicating such concentration or temperature is critical. *In re Aller*, 220 F.2d 454, 456, 105 USPQ 233, 235 (CCPA 1955).

With respect to claim 32, modified Makrides '846 further discloses that the metals from Group V-B (Nb, Ta, and V) have a thickness of 0.001 inch (25.4 μm) and the palladium films have a thickness each of 1000 angstroms (0.1 μm) (col. 4, lines 30-35), giving a total palladium membrane thickness of 25.4 μm.

16. Claims 20 and 24-25 are rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846), Lee et al (US Patent No. 6,379,524), and Stevens et al (USPGPub 2003/0141018) as applied to claims 18 and 19, and in further view of Steinberg et al (4,055,686).

With respect to claims 20 and 24-25, the references are cited as discussed for claims 18 and 19. Modified Lee '524 further teaches methods of prepping (i.e. activating) a stainless steel support in a temperature range of 4°C (col. 5, lines 40-45) and 800°C (col. 6, lines 10-15). However modified Lee '524 is limited in that while it is discussed to activate the support, it is not suggested to polish the substrate holder.

Steinberg '686 teaches a method of forming metal hydride films on a surface of a substrate that rests on a stainless steel shroud which is on top of the substrate holder (abstract; col. 2, lines 65-67). Steinberg '686 further teaches that metal hydride films are used in hydrogen diffusion applications (col. 1, lines 33-36). Polishing of the substrate, and thus stainless steel shroud, is by mechanically polishing with number 600 emery paper (col. 4, lines 43-45) followed by cleaning with ultrasonic bath (col. 4, lines 58-59). Etching (i.e. acid-washing) with 10% HCl aqueous solution is also discussed (col. 3, lines 52-53) along with sputter etching (i.e. electro-polishing) via DC and radio frequency sputter etching after filling the substrate, and therefore support, with a metal hydride (col. 2, lines 29-59). Steinberg '686 cites the advantage of cleaning as providing a substrate system tolerant to a difference between the coefficients of expansion of the film and substrate due to a strong bond between the film and substrate (col. 2, lines 12-19).

It would have been obvious to one of ordinary skill in the art to use the polishing and cleaning methods taught in Steinberg '686 for modified Lee '524 in order to gain the advantages of tolerance in differences of coefficients of expansion between the film and substrate.

17. Claim 23 is rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846), Lee et al (US Patent No. 6,379,524) and Stevens et al (USPGPub 2003/0141018) as applied to claim 22, and in further view of Kimura (US Patent No. 4,574,056).

With respect to claim 23, the references are cited as discussed for claim 22. However modified Makrides '846 is limited in that it is not suggested to mix the metal powder with a palladium paste and epoxy resin.

Kimura '056 teaches electroconductive pastes that are used to bond semiconductor elements (i.e. metals) onto substrates (abstract; col. 1, lines 18-21). Furthermore, Kimura '056 discusses electroconductive pastes known in the art include those produced by blending fine electroconductive powders of palladium and heat-resistant binders such as epoxy resin (col. 2, lines 58-63). Kimura '056 cites the advantage of using this paste as not requiring any undercoat electrode treatment (col. 1, lines 34-37).

It would have been obvious to one of ordinary skill in the art to use the palladium paste and epoxy resin taught in Kimura '056 for the membrane of modified Makrides '846 in order to gain the advantage of not requiring an undercoat electrode treatment for the substrate and layers.

18. Claims 26-27 is rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846), Lee et al (US Patent No. 6,379,524), and Stevens et al (USPGPub 2003/0141018) as applied to claim 18 above, and further in view of Harkness (US Patent No. 6,265,086).

With respect to claims 26-27, the references are cited as discussed for claim 18. However modified Makrides '846 is limited in that while it is disclosed that a silver-palladium alloy is electroless plated, the salt solution is not specified.

Harkness '086 teaches a method for selective electroless metal deposition on a substrate (abstract). Harkness '086 further teaches suitable sources of metal ions include salts of metals, such as silver nitrate and palladium acetate, with a concentration in the range of about 0.001 to about 10 weight percent (col. 6, lines 20-29). Furthermore, Harkness '086 states that "electroless plating solutions also often contain solvents for the metal salt, suitable reducing agents, bases, complexing agents to solubilize the metal salt and special additives to control the solution stability and plating rate (col. 6, lines 30-33). Harkness '086 suggests a reducing agent of hydrazine (NH_2NH_2) with a concentration range of about 0.01 to about 10 weight percent (col. 6, lines 38-39), a base of ammonium hydroxide (NH_4OH) with a concentration of 0.01 to about 10 weight percent (col. 6, lines 45-46), and a complexing agent of EDTA with a concentration range of about 0.01 to about 10 weight percent (col. 6, lines 52-54).

It has been held that differences in concentration or temperature will not support patentability of subject matter encompassed by the prior art unless there is evidence indicating such concentration or temperature is critical. *In re Aller*, 220 F.2d 454, 456, 105 USPQ 233, 235 (CCPA 1955).

It would have been obvious to one of ordinary skill in the art to use a solution of compounds taught by Harkness '086 for the solution of modified Makrides '846 in order to gain the advantages of controlling the solution stability and plating rate.

19. Claim 30 is rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846), Lee et al (US Patent No. 6,379,524), and Stevens et al (USPGPub 2003/0141018) as applied to claim 18, and in further view of Gopalraja et al (USPGPub 2004/0094402).

With respect to claim 30, the references are cited as discussed for claim 18. However modified Makrides '846 is limited in that while sputtering is disclosed, specific vacuum pressures, power input, and temperature are not specified.

Gopalraja '402 teaches a DC magnetron sputtering (abstract) for a palladium (Pd) and silver (Ag) (p. 3, para 0016) while also suggesting electrochemical plating (i.e. electroless plating) and electroplating (p. 3, para 0021-0022). Furthermore, Gopalraja '402 discusses suitable pressure ranges as 1-30 mTorr or 1-5 mTorr (p. 5, para 0043) accompanied with temperatures between 50 to 70°C (p. 2, para 0014). Gopalraja '402 also discusses a power source capable of biasing a pedestal, where the power range is preferred from 150 to 300 W (p. 5, para 0053). Gopalraja '402 cites the advantage to sputtering using these parameters as confining the plasma generated by capacitive coupling to increase the plasma density, leading to an increased ionization rate in addition to the process being promoted at low pressure (p. 3, para 0024-0025).

It would have been obvious to one of ordinary skill in the art to use the parameters specified for a DC sputtering magnetron taught in Gopalraja '402 for the sputtering apparatus of modified Makrides '846 in order to gain the advantages of superior ionization and improved process at lower pressures.

It has been held that in the case where the claimed ranges "overlap or lie inside ranges disclosed by the prior art" a *prima facie* case of obviousness exists. *In re Wertheim*, 541 F.2d 257, 191 USPQ 90 (CCPA 1976). .

20. Claim 31 is rejected under 35 U.S.C. 103(a) as being unpatentable over Makrides et al (US Patent No. 3,350,846), Lee et al (US Patent No. 6,379,524), and Stevens et al (USPGPub 2003/0141018) as applied to claim 18 above, and further in view Harris (US Patent No. 4,313,013).

With respect to claim 31, the references are cited as discussed for claim 18. However modified Makrides '846 is limited in that while the membrane is heated, it is not suggested at what temperature, for how long, in what type of atmosphere, or what percentage of hydrogen comprises the gas stream.

Harris '013 discloses a palladium or palladium alloy hydrogen diffusion membrane (abstract) and method of treating (i.e. annealing) the membrane with heat for a specific time in a specific atmosphere (col. 2, lines 53-67). Harris '013 states that silver is used an alloy with palladium (col. 2, lines 26-32). Temperatures of between 350°C and 450°C are used at a time period between 0.1 and 10 hours (col. 2, lines 64-67). Harris '013 also states that hydrogen, argon, or nitrogen gas are used, either static (i.e. atmosphere) or flowing. Furthermore, Harris '013 conducts an example of this with the argon stream being analyzed to determine its content, which is depicted in Table 1 (col. 3, lines 64-68). Table 1 depicts a hydrogen content in the range of 6.6-13.0. Harris '013 cites the advantage of the treatment as a production and recovery of hydrogen from a hydrocarbon source (col. 1, lines 43-47).

It would have been obvious to one of ordinary skill to use the treatment method taught in Harris '013 for the manufacturing method of modified Makrides '846 in order to gain the advantages of production and recovery of hydrogen from a hydrocarbon source.

It has been held that in the case where the claimed ranges "overlap or lie inside ranges disclosed by the prior art" a *prima facie* case of obviousness exists. *In re Wertheim*, 541 F.2d 257, 191 USPQ 90 (CCPA 1976).

21. Claim 33 is rejected under 35 U.S.C. 103(a) as being unpatentable over Lee et al (US Patent No. 6,379,524) in view of Stevens et al (USPGPub 2003/0141018).

With respect to claim 33, Lee '524 discloses a process for manufacturing a composite membrane for separation of hydrogen gas using palladium (abstract). Lee '524 further discloses examples of plating methods designed to prepare a palladium composite membrane by means of coating (i.e. filling) palladium metal membrane and palladium alloy membrane on a porous support by electroless plating (col. 2, lines 17-20), with a porous support derived from stainless steel (col. 2, lines 11-13). It is known that electroless plating a palladium metal involves using a palladium salt solution, as evidenced by Nogami et al (US Patent No. 6,059,940; col. 1, lines 29-33). Furthermore, Lee '524 discusses preparing an alloy using palladium and transition metals, such as silver (col. 3, lines 57-61). However Lee '524 is limited in that while the membrane is heated, it is not specified whether the membrane is slowly cooled afterward.

Stevens '018 teaches an electroless deposition apparatus comprising at least one metal selected from the group consisting of noble metals, semi-noble metals, alloys

therof (abstract), with examples of noble metals being silver and palladium (p. 2, para 13). Furthermore, Stevens '018 discusses that the process of depositing the conductive (i.e. palladium-silver alloy) layer (i.e. membrane) may include annealing the substrate in a thermal anneal chamber. Stevens '018 states the advantage of annealing is enhancement of the properties of the deposited materials (p. 10, para 88).

It would have been obvious to one of ordinary skill in the art to include an anneal step taught in Stevens '018 for the process of Lee '524 in order to gain the advantage of enhanced properties of the palladium-silver membrane.

Conclusion

24. The prior art made of record and not relied upon is considered pertinent to applicant's disclosure. US Patent No. 7,112,287; US Patent No. 5,215,729; USPGPub 2004/0087248.

25. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Michael Band whose telephone number is (571) 272-9815. The examiner can normally be reached on Mon-Fri, 8am-4pm, EST.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Alexa Neckel can be reached on (571) 272-1446. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

26. Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR.

Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

MAB

ALEXA D. NECKEL
SUPERVISORY PATENT EXAMINER